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Determination of the diffusion length and second spatial moment of thermal neutrons in water with a small boron chamber

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With 11 figures in the text

A. Determination of the diffusion length

I. Introduction

The diffusion length L of thermal neutrons in a moderator is defined as the average distance thermal neutrons will diffuse in the moderator from the point where they become slowed down to thermal energies to the point where they get captured. The quantity L according to diffusion theory is related to the other fundamental constants of the moderator by the following equation:

$$L^2 = \frac{\lambda_t \cdot \Lambda}{3} \tag{1}$$

where λ_t = transport mean free path
Λ = absorption mean free path.

According to the open literature the diffusion length for water has earlier been experimentally determined by GAMERTSFELDER and GOLDHABER [1] and by BERTHELOT et al [2]. The authors (1) report a value $L = 3$ cm ± 10 % and (2) 2.77 ± 0.04cm. As all natural neutron sources give fast neutrons, the method used by the authors cited above essentially has been the same, to bring about a differential neutron source. Since the determination (2) of L has been done with a Mn-detector and for a special case it seemed very interesting to make an investigation under more general conditions.

II. The method

The diffusion equation for thermal neutrons is simplified in the stationary case and when the slowing down density $= 0$ to

$$\Delta \varrho - \frac{\varrho}{L^2} = 0 \tag{2}$$

where ϱ = density of thermal neutrons.

The solution of this equation for a point source in an infinite medium is

$$\varrho(r) = \text{const.}\ \frac{e^{-\frac{r}{L}}}{r} \tag{3}$$

where r = distance from the source of thermal neutrons.

To bring about a source of purely thermal neutrons a differential measurement is made. At first the spatial distribution in pure water of the thermal neutrons originating from the slowing down of fast neutrons from a Ra-α-Be-source is studied. Then an absorber (Cd) of thermal neutrons which is spherically symmetric in relation to its source is introduced, and the spatial distribution of the thermal neutrons outside the absorber is studied. The difference between these two measurements represents the density of thermal neutrons captured by the absorber. This density will satisfy the equation (3).

Throughout this investigation the term "thermal neutrons" is used. As this term is used in a rather diffuse way in the literature it is worth while mentioning that a better term covering the experimental conditions in this experiment would be C-neutrons, a term due to Fermi. In reality it is not possible to state that only thermal neutrons in the strict sense are measured in differential experiments of this type. The correct statement is that the energy of the measured neutrons is below the Cd-absorption limit. Below this limit, the energy spectrum of the neutrons is unknown and is in fact strongly disturbed by the absorbing cadmium medium, as will be shown by the measurements.

III. Experimental arrangement

Two neutron sources were used, 100 mC of Ra mixed with 500 mg Be and 250 mC Ra mixed with 1250 mg Be, contained in cylinders of monelmetal, the external dimensions of which were 35 mm long, 14 mm wide, and 40 mm long,

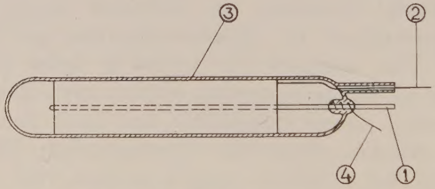

Fig. 1. The boron chamber. Total length 30 mm, diameter 7 mm. Both electrodes of platinum. 1. Collecting electrode. 2. High tension wire. 3. Cylinder. 4. Guard ring.

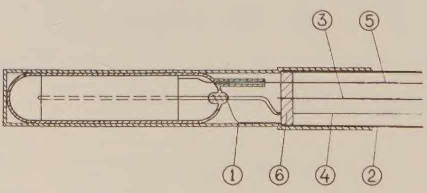

Fig. 2. Mounting of the boron chamber at the end of the coppertube. 1. Polysterene tube. 2. Coppertube. 3. Copperfoil. 4. Collecting wire. 5. High tension wire. 6. Polysterene plug.

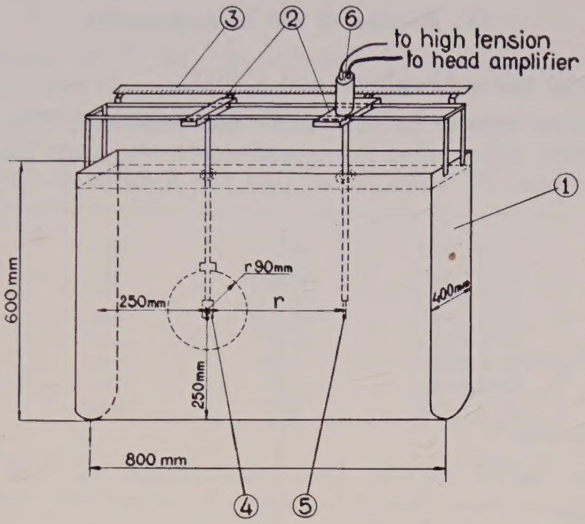

Fig. 3. Experimental arrangement. 1. Watertank with steel rails. 2. Carriages. 3. Steel scale. 4. Ra-α-Be-source (the spheres indicated by dots). 5. Boron chamber. 6. Pre-amplifier.

17 mm wide, respectively. In order to bring forth a differential neutron source, two spheres were manufactured, each of 90 mm radius, one made of 0.4 mm Al-sheet, the other of 0.7 mm Cd-sheet. With a thin rod of polysterene, at one end furnished with a small cylindrical holder, the sources could be fixed with their centres in the centres of the spheres. The other end of the rod was rigidly connected to a carriage, movable along two iron rails over a U-shaped watertank of stainless steel, the dimensions of which were: 80 cm long, 40 cm wide, and 60 cm deep. During the measurements this tank was filled with 140 liters of distilled water. As detector a small boron chamber was used, the construction of which is seen in Fig. 1. The chamber was filled to 1 atm. with BF_3. It was fixed to one end of a thin coppertube which by a copperfoil was divided into two halves containing high tension and collecting wires, respectively. The other end of the tube was rigidly connected to another carriage which was also movable along the rails. Both carriages had a pionter showing directly on a steel scale the distance between the centre of the source and the centre of the boron chamber. The chamber as well as the copper tube were surrounded by a thin polysterene tube to prevent water from penetrating into the tube containing the wires (Fig. 2). Directly on the carriage holding the boron chamber a pre-amplifier was fixed from which cables led to the head amplifier (total amplification of the primary impulse $\sim 10^6$) and a scale of 64 with discriminator. The whole arrangement is shown in Fig. 3. With this apparatus two main series of measurements were made. First, the neutron density was measured with the Al-sphere filled with solutions of $CdSO_4$ in varying concentrations. Neutron source 100 mC Ra-α-Be. Secondly the density was studied outside the waterfilled Cd-sphere. Neutron source 250 mC Ra-α-Be.

IV. Results of the measurements

A. Measurement with Al-sphere and $CdSO_4$

First the neutron density in pure water was measured. The same number of pulses was counted either when the measurement was made with the waterfilled Al-sphere or without it, which shows that the disturbance in neutron density caused by the thin sphere was negligeable. Then the sphere was filled with 10, 20, 37, 50, 62, 100, 150, and 200 g $CdSO_4$ dissolved in the 3 litres of water contained in the sphere. The result, represented by the group of curves, is shown in Fig. 4. If the difference between the density in pure water and the density outside the sphere, as measured point by point, is represented by $\varrho(r)$, it will be seen that $r \cdot \varrho(r) = \text{const. } e^{-\frac{r}{L}}$ according to theory shall form a line when plotted on semilogaritmic paper. In Fig. 4 this line is shown for the highest end lowest concentrations of Cd in the sphere. The values of L for the various concentrations of Cd calculated by the method of least squares are given in table 1.

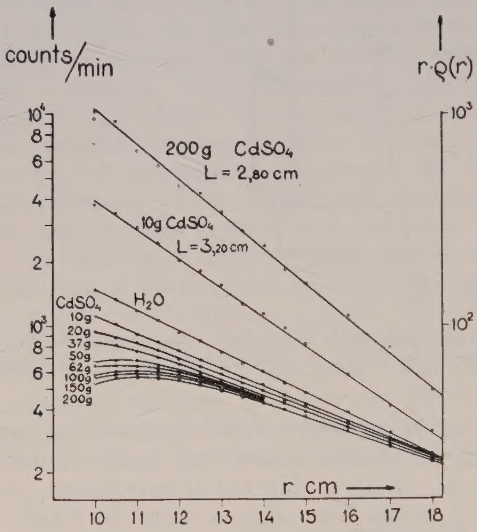

Fig. 4. Neutron density curves measured in pure water and outside the Al-sphere with Cd-solutions. The two upper curves give $r \cdot \varrho(r)$ as a function of r.

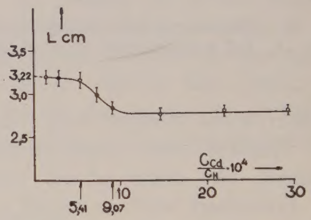

Fig. 5. The diffusion length as a function of the ratio of concentration of cadmium to hydrogen in the sphere.

L, thus, becomes a function of the Cd-concentration in the sphere as is graphically shown in Fig. 5.

This dependence of L on the Cd-concentration must be intimately connected with the idealization made in the interpretation of the experimental results. Equation (3) predicts the neutron density distribution from a point source of thermal neutrons but in this experiment a sphere is substituted for the point source. As will be seen, there are at least two possibilities of making systematical errors in these measurements.

Table 1

Experimental values of L for the different Cd-concentrations in the sphere. C_{Cd} and C_H atomic concentrations of Cd and H.

g $CdSO_4$	$\frac{C_{Cd}}{C_H} \cdot 10^4$	L cm
10	1.46	3.20 ± 0.07
20	2.92	3.18 ± 0.09
37	5.41	3.16 ± 0.09
50	7.32	2.99 ± 0.08
62	9.07	2.84 ± 0.07
100	14.6	2.76 ± 0.07
150	22.0	2.80 ± 0.06
200	29.3	2.80 ± 0.06

a. In order that the measurements shall give the correct value of L, it is necessary that the distribution of sources of thermal neutrons outside the sphere at all points and at all Cd-concentrations is the same as the source distribution in pure water. Otherwise the density $\varrho(r)$ would be erroneous, since in taking the difference of the two densities with and without Cd-solution it is anticipated that nothing is altered in the neutron distribution except that a certain percentage of thermal neutrons is absorbed in the sphere.

b. Secondly, a great error may be caused by the large extension of the source. Neutrons slowed down to thermal energies in the neighbourhood of the sphere but outside of it have great chances to diffuse back into the sphere and get captured in Cd; in the case of pure water, on the other hand, these neutrons are able to diffuse back into the region where they can be counted. The sphere thus excerts a shadowing effect upon neutrons slowed down in its immediate neighbourhood. This effect must depend upon the solid angle occupied by the sphere as seen from the point of measurement. Thus, it contributes to bring about an erroneous reduction in neutron density in the vicinity of the sphere. It is to be expected that this error is considerable up to a distance of a diffusion length from the surface of the sphere and at high Cd-concentrations.

B. Investigation of the source distribution at various conditions

Since it is impossible to measure the source distribution for thermal neutrons directly, the only possibility to get an idea of this is to measure the distribution of resonance neutrons with a detector of a material having a sharp resonance immediately above the thermal energy region. For this indium was chosen which has a pronounced resonance peak at 1.44 eV[3] and the absorption cross section of which is very high, ~ 10 000 barns, so that a reasonably high activity is induced even at low neutron densities. With In-detectors (25 × 25 mm, 48 mg/cm^2) screened by 0.3 mm Cd-plates, the spatial distribution was measured outside the Al-sphere filled with pure water, 10 g $CdSO_4$ and 150 g $CdSO_4$ in solution. The result of these measurements is graphically shown in Fig. 6. As will be seen, the density of these neutrons at the higher Cd-concentration

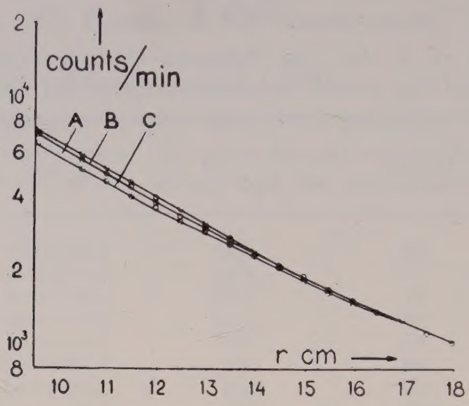

Fig. 6. Density of In-resonance neutrons outside the Al-sphere. A. Pure water. B. 10 g $CdSO_4$ in the sphere. C. 150 g $CdSO_4$ in the sphere.

in the sphere differs markedly from the density in pure water. At $r = 10$ cm the density is lowered by some 10 %, which decrease does not vanish until $r \sim 17$ cm. At the lower Cd-concentration the deviation is about 3 % at $r = 10$ cm and vanishes at $r \sim 14$ cm. The reason for this decrease in the density of In-resonance neutrons might be that the absorption cross section of Cd is approximately 50 times the absorption cross section of H for neutron energies > 1.5 eV $\left(\frac{C_{Cd}}{C_H} \cdot \frac{\sigma_{Cd}}{C_H} \sim 0.1 \text{ for } \frac{C_{Cd}}{C_H} \sim 22 \cdot 10^{-4} \text{ and } \sim 0.03 \text{ for } \frac{C_{Cd}}{C_H} \sim 1.5 \cdot 10^{-4}, \text{ both relations giving } \frac{\sigma_{Cd}}{\sigma_H} \sim 50\right)$, or else Cd might have less pronounced resonances for higher neutron energies. The perturbation in the source distribution decreases with Cd-concentration. The result of this perturbation is that the thermal neutron density is more decreased near the sphere than at some distance from it, and thus $\varrho(r)$ is increased in the same manner which tends to give a too low value of L, especially at high Cd-concentrations.

C. A study of the shadowing effect

In order to get a value of L under different absorption conditions and, if possible, to show the second disturbance — the shadowing effect — more clearly, a series of measurements were made with the Cd-sphere. As in the first experiment (A), the density distribution of thermal neutrons was measured in pure water and outside the waterfilled Cd-sphere. The density curves are shown in Fig. 7. The curve $\varrho(r) \cdot r = \text{const.}\ e^{-\frac{r}{L}}$, given in Fig. 8, has a peculiar appearance. At $r \sim 13.5$ cm the line is broken. For $10 \leq r \leq 13.5$ cm the line has a greater slope than for $r > 13.5$ cm. But this must be the case if the shadowing effect is considerable, because then the density in the neighbourhood of the sphere undergoes an extra decrease, which appears as an increase in $\varrho(r)$ and consequently a greater slope of the line. Furthermore, it is seen that this nick of the curve occurs at a distance from the sphere of ~ 45 mm,

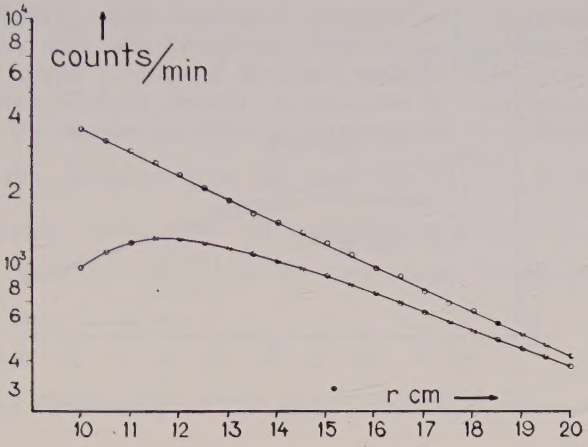

Fig. 7. Neutron density in pure water and outside the Cd-sphere.

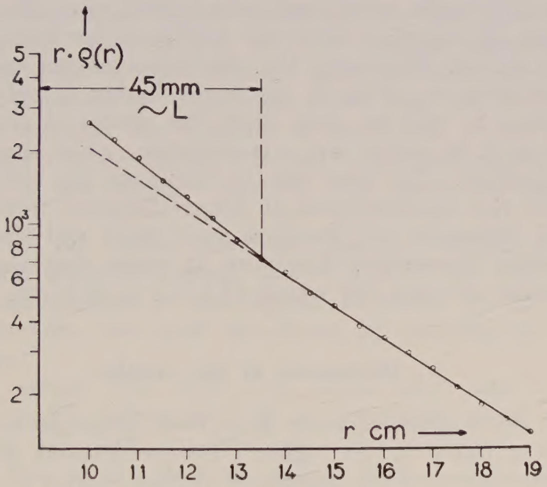

Fig. 8. The line giving L. The nick on the line occurs at approximately one diffusion length from the surface of the sphere.

which approximately equals the diffusion length. In order to determine L, only points for which $r \geq 13.5$ cm are used. The method of least squares gives

$$L = 3.29 \pm 0.09 \text{ cm}$$

a value which is in good agreement with the value 3.20 cm received at the lowest Cd-concentration in the first experiment (A).

In order to further investigate the shadowing effect caused by the Cd-sphere another measurement was made. With empty spheres the neutron density was measured outside both the Al-sphere and the Cd-sphere. As is shown in Fig. 9, a very strong depression in neutron density is caused outside the Cd-sphere.

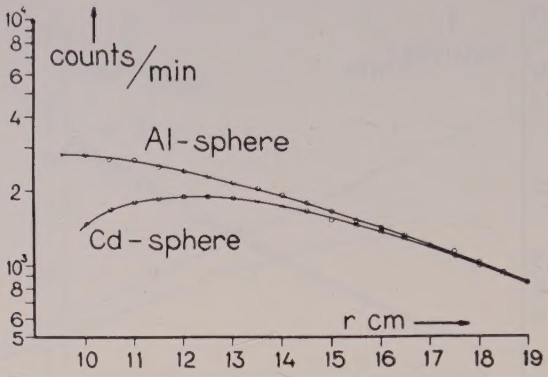

Fig. 9. Neutron density outside empty spheres.

At $r = 10$ cm the decrease amounts to ~ 50 %, at $r = 13.5$ cm it amounts to ~ 10 % and at $r = 18$ cm it is ~ 1 %. In the case when the sphere is empty, however, the effect is much more pronounced than when the sphere contains a moderator, because all neutrons from the source in the former case are slowed down outside the sphere. Moreover, the absorption of the sphere is noticeable at larger distances from it, owing to the much higher density of thermal neutrons. In any case it will be seen that the source of error caused by the shadowing effect must be taken into consideration, when the goal is to determine L by this method. The best way to eliminate the two errors now mentioned, is to make the measurements at larger distances from the sphere. But at $r > 20$ cm the difference $\varrho(r)$ becomes very small and the statistical errors make a measurement impossible. Therefore, it seems that the best value of L is obtained if points are used for which $13.5 \leq r \leq 19.5$ cm.

V. Discussion of the results

Now that the two sources of error have been investigated, it is possible to discuss the curve in figure 5. At higher Cd-concentrations in the sphere practically all neutrons slowed down outside of it that happen to diffuse back into the sphere will be absorbed, and the perturbation in thermal sources is rather constant. In that region the L-value will be constant. At a certain concentration ratio, $\frac{C_{Cd}}{C_H} \sim 9.10^{-4}$, the shadowing effect will begin to decrease because some thermal neutrons can diffuse back out of the sphere and get counted, which means that $\varrho(r)$ decreases comparatively much in the neighbourhood of the sphere and the value of L increases. When $\frac{C_{Cd}}{C_H} \sim 5 \cdot 10^{-4}$, the absorption probability in Cd then being only 5 times the absorption probability in H, the neutrons are able to diffuse to and from through the wall of the sphere and have a greater chance of being counted. Thus, the value of L will increase only slowly with the Cd-Concentration for $\frac{C_{Cd}}{C_H} < 5 \cdot 10^{-4}$. Even at low concentrations of $CdSO_4$, there remains a disturbance in the source distribution. The measurement with the Cd-sphere, where this disturbance is inappreciable for $r > 13$ cm, gives a value of L which must be very probable on the assumption that the neutron density is measured at such large distances from the sphere that the shadowing effect is inappreciable or eliminated. The measurement with the Al-sphere gives the limit $\lim_{C_{Cd} \to 0} L \left(\frac{C_{Cd}}{C_H}\right)$, which must be another very probable value, since all perturbations in neutron density vanish with C_{Cd}. The curve $L = f\left(\frac{C_{Cd}}{C_H}\right)$ is for that reason extrapolated to $C_{Cd} = 0$ giving $L = 3.22$ cm.

VI. Discussion of the other experimental errors

With the value of L measured as outlined above one may assume that the two greatest systematic errors are eliminated or very small.

A third systematic error might be caused by the finite size of the boron chamber. This would tend to give too high neutron densities. On the other hand, a small fraction of the neutrons must get absorbed in the glass walls of the chamber. Moreover, a calculation shows that the absorption of thermal neutrons by the boron in the chamber is equivalent to the absorption in the corresponding water volume ($N_H \sigma_H \sim N_B \sigma_B$). For these reasons the error caused by the finite size of the detector must be very small. No correction is applied for it.

The counting of the boron chamber, due to neutrons of energies greater than the cadmium absorption limit, was < 1 % of the total counting. Moreover, since the distribution of non-thermal neutrons is little disturbed by introducing the absorbers, the error introduced by counting non-thermal neutrons must be negligible.

The statistical errors were kept very small, since least 10 000 pulses were always counted, which corresponds to a statistical error of 0.5—1 %. The background count was 0.04 impulses/min. The statistical uncertainty in $\varrho(r)$ thus amounts to 2 % at $r = 10.0$ cm and to 5 % at $r = 18.5$ cm. During the whole series of measurement the reproducibility of counting was checked and all measurements were made several times. The measured number of pulses always fell within the statistical limits of error, which guarantees the reliability of the electronic equipment.

The errors in r amounts to ± 0.4 mm, the relative error in r thus amounts to 0.4 % at most.

The theory of this experiment requires a slowing down medium of infinite size, a condition which is not satisfied. When, however, only thermal neutrons are studied and the detector never approaches the walls of the tank closer than six diffusion lengths, this disturbance in boundary conditions is neglible.

The total uncertainty in L is estimated to be 4 %.

The measurements described above then give as a weighted mean

$$L = 3.25 \pm 0.13 \text{ cm}$$

which means, correctly speaking, that the diffusion length of C-neutrons under the experimental conditions specified above is 3.25 cm, this value being corrected for the disturbances in the energy distribution of the C-neutrons that arise during measurements. This value thus agrees with the value reported by GAMERTSFELDER and GOLDHABER [1] within limits of error. The value 2.77 ± 0.04 cm reported by BERTHELOT et al [2] agrees with the value 2.80 ± 0.06 found in the measurements with the Al-sphere at higher Cd-concentrations. This value, however, is not corrected for the two great disturbances in the neutron distribution as shown in these measurements.

Furthermore the diffusion theory approximation suffers from a theoretical error, which is a function of the ratio $\frac{\sigma_a}{\sigma}$ (σ_a = absorption cross-section, σ = total cross-section). [8] This error might be estimated to ~ 1 % in the mathematical description of the density of neutrons in water. A better approximation in place of eq. 1, for instance, is

$$L^2 = \frac{1}{3} \Lambda \cdot \lambda_t \left(1 + \frac{4}{5} \frac{\lambda_t}{\Lambda}\right),$$

this expression resulting from the general Bolzmann equation. [8] This theoretical error in ordinary diffusion theory of course increases the uncertainty in the value of L given above.

B. Determination of the second spatial moment

I. Introduction and theory

By age-diffusion theory [4, 5] the second spatial moment $\langle r^2 \rangle$ for thermal neutrons is functionally related to the diffusion length by the following relation

$$\frac{\langle r_{th}^2 \rangle}{6} = M_{th}^2 = \tau_{th} + L^2 \tag{4}$$

where M_{th}^2 = migration area
τ_{th} = "age" of thermal neutrons
L = diffusion length of thermal neutrons.

In spite of the fact that age-diffusion theory is a poor approximation in problems of slowing down in a hydrogenous moderator like water — especially when the neutrons have so high initial energies as Ra-α-Be-neutrons — it seemed interesting to determine $\langle r_{th}^2 \rangle$. In fact this measurement with a small boron chamber might give an interesting comparation with results received in measurements with foils by several authors. [6, 7].

Experimentally the density $\varrho(r)$ of thermal neutrons is measured as a function of the distance r from the source. The total number of neutrons counted by the boron chamber is then

$$N = 4\pi \int_0^\infty I\, r^2\, dr$$

where I is the number of counts/min at r.

The mean square of the migration distance is then found from

$$\langle r_{th}^2 \rangle = 6\, M_{th}^2 = \frac{\int\limits_0^\infty I\, r^4\, d\, r}{\int\limits_0^\infty I\, r^2\, d\, r}.$$

II. Experimental equipment and results

The whole apparatus used to determine $\langle r^2 \rangle$ is the same as described above in the determination of L. The density of thermal neutrons was measured from $r = 4$ cm to $r = 38$ cm. For $4 \leq r \leq 20$ cm the 100 mC Ra-α-Be-source was used. For $12 \leq r \leq 38$ cm the 250 mC neutron source was used in order to get reasonably high counting rates. In the region $12 \leq r \leq 20$ cm the two sources were calibrated against each other. The result of this calibration is given in the table.

Table 2

Calibration of the 250 mC source against the 100 mC source.

r cm	I counts/min 100 mC	I counts/min 250 mC	$\frac{I_{100}}{I_{250}}$
12.0	947	2173	0.436
13.0	758	1720	0.440
14.0	611	1385	0.441
15.0	490	1123	0.436
16.0	398	911	0.438
17.0	321	734	0.437
18.0	269	614	0.438
19.0	216	490	0.441
20.0	179	415	0.432
		Mean value	0.438

The readings for $r > 20$ cm with the 250 mC source were adjusted to the proper values corresponding the 100 mC source by multiplication with the factor 0.438. The electronic apparatuses run continously during the course of the experiment, and no drift could be detected within the statistical accuracy of the counts. At each point up to $r = 25$ cm 10 000 to 20 000 pulses were counted. The least number of pulses counted for $r > 25$ cm was actually 7000. The statistical accuracy thus amounts to ~ 1 %. In Fig. 10 the function $I\, r^2 = f(r)$ is shown on semilogarithmic scale. This curve shows an exponential decrease when $r > 20$ cm with the relaxation length 9.93 cm. Assuming an exponential decrease without limit with this relaxation length, points on the curve $I\, r^2 = f(r)$ were calculated for $r > 38$ cm and $I\, r^4$ was formed. However, $I\, r^4 = \varphi(r)$ shows no clear exponential decrease until $r > 86$ cm (Fig. 11). The space integral $(I\, r^2\, d\, r)$ was evaluated by formal integration for $r > 38$ cm, assuming exponential decrease without limit, and by Simpsons rule over the

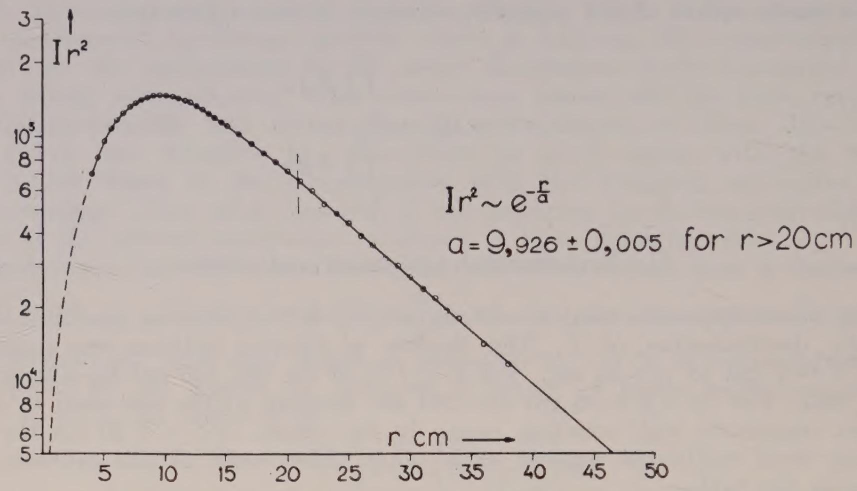

Fig. 10. Semilog plot of $I\,r^2 = f\,(r)$, the curve showing exponential decrease from $r = 20$ cm.

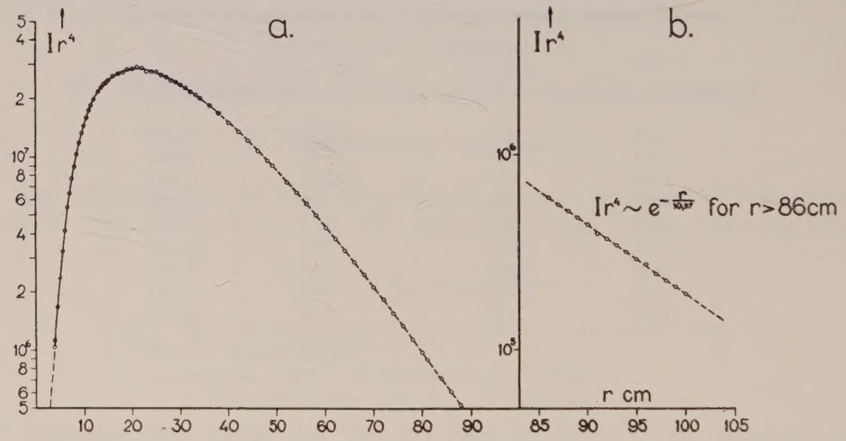

Fig. 11. Semilog plot of $I\,r^4 = \varphi\,(r)$. The dotted part of the curve is calculated from the exponential decrease of $I\,r^2 = f\,(r)$. The curve $I\,r^4 = \varphi\,(r)$ shows no safe exponential decrease until $r \geq 86$ cm.

remainder. $(I\,r^4\,d\,r)$ was evaluated by Simpsons rule in the region $0 < r \leq 86$ cm and by formal integration over the remainder. The result is:

$$\langle r_{th}^2 \rangle = \frac{\int\limits_0^\infty I\,r^4\,d\,r}{\int\limits_0^\infty I\,r^2\,d\,r} = \frac{973.4 \cdot 10^6}{266 \cdot 10^4} = 366 \text{ cm}^2.$$

The corresponding migration area is

$$M^2 = \frac{\langle r^2 \rangle}{6} = 61.0 \text{ cm}^2.$$

The above value of L inserted in equation 4 gives

$$\tau_{th} = (61.0—10.8) \text{ cm}^2 = 50.2 \text{ cm}^2.$$

This value of $\langle r_{th}^2 \rangle$ is some 10 % higher than the value reported by Rush [6] and by Munn and Pontecorvo [7]. As is easily seen there are to be expected relatively large discrepances between this measurement with boron chamber and the measurements [6, 7] with In- and Dy-detectors respectively. With Dy-detectors, for instance, not only the thermal neutron distribution is measured, but the distribution of resonance neutrons is superposed, because the cross section of Dy shows disturbing resonances at a few eV^3. This tends to give lower values of $\langle r_{th}^2 \rangle$. The same reasoning holds for In-detectors. Boron, on the other hand, is a strict $\frac{1}{v}$-absorber practically over the whole energy region of the Ra-α-Be-neutrons. As measurements show, the counting rate of the Cd-shielded BF_3chamber is < 1 % of the unshielded chamber, so that the error introduced by counting non-thermal neutrons is very small in the case of a boron-chamber.

Acknowledgements

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